

# Outdoor/indoor relationships of PM<sub>1</sub> and black carbon: sources and infiltration processes in an urban environment

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## Abstract

Ambient air emissions of sub-micron particles infiltrate into indoor environments and play a major role in indoor air quality. Discriminating between particles of indoor vs. outdoor origin is therefore essential when assessing indoor air pollutant levels and characteristics. Outdoor/indoor relationships of traffic-derived nanoscaled particulates (PM<sub>1</sub> and black carbon) were determined in a typical Mediterranean environment. Results evidenced the major impact of outdoor vehicular traffic emissions on indoor particulates: despite windows remaining closed at all times, 70% of indoor BC and 73% of indoor PM<sub>1</sub> originated from outdoor emissions. This was probably due to inadequate insulation of the building. Outdoor/indoor penetration ratios were relatively constant for BC ( $1.29 \pm 0.08$ ) but not for PM<sub>1</sub> ( $1.95 \pm 0.38$ ), suggesting it is advisable to monitor the variability of penetration factors over time. Particle infiltration seemed to depend not only on physical barriers (building envelope, ventilation systems, etc.), but also on the physico-chemical properties of the particulates. Printing and photocopying contributed with 25-30% ( $546 \text{ ng/m}^3$ ) of total indoor BC. Dust re-suspension by worker passage was the main indoor source of PM<sub>1</sub> (15-20%,  $1.1 \text{ } \mu\text{g/m}^3$ ).

**Keywords:** exhaust, infiltration, penetration ratio, Barcelona, Spain, shift analysis.

## Introduction

Numerous research works available in the literature have reported significant health risks associated with exposure to particulate matter (PM) (Brunekreef et al., 1997; Pekkanen 1997; Lippmann & Schlesinger, 2000; Künzli et al., 2000; Pope & Dockery, 2006). In recent years research interest has been directed towards fine and ultrafine particles (particulates with aerodynamic diameter  $<1 \mu\text{m}$  and  $<0.1 \mu\text{m}$ , respectively). Epidemiological studies (Donaldson et al., 2001; Oberdoster, 2001) indicate that fine and ultrafine particles may have a greater potency to cause adverse health effects than larger particles, due to their increased deposition rates in the lower respiratory tract (Jaques & Kim, 2000) and their higher transition

1 metal content (Shi et al., 2004). Furthermore, ultrafine particles (UFP) have also been  
2 suggested to be more toxic because of the large surface area available for biologic  
3 interactions with lung cells (Chio et al., 2008). There is little information on the trend in UFP  
4 in European urban atmospheres, but the increased load of diesel vehicles and recent data  
5 (Mejia et al., 2007) suggest an upward trend. Recent works have shown that traffic is the  
6 origin of 90% of the UFP load in urban environments (Pey et al., 2009).

7  
8 Population exposure to both supra-micron and nanoscaled particulate concentrations is  
9 usually considered to take place in outdoor environments. However, it is estimated that on  
10 average adults spend 60-90% of their time indoors (Klepeis et al., 2001). Whereas data on  
11 outdoor fine and ultrafine particle levels and physico-chemical characteristics are widely  
12 available in Europe (Putaud et al., 2004; Querol et al., 2004; Hueglin et al., 2005; Andersen  
13 et al., 2007; Lonati et al., 2007), fewer data are available in the literature regarding the levels  
14 and characteristics of atmospheric particulates in indoor air. However, the interest on this  
15 topic is increasing in the US (Jones et al., 2000; Geller et al., 2002; Sawant et al., 2004;  
16 Polidori et al., 2006; Lunden et al., 2008; Martuzevicius et al., 2008), Europe (Fischer et al.,  
17 2000; Kingham et al., 2000; Cyrus et al., 2004; Hänninen et al., 2004; Hoek et al., 2008)  
18 and Asia (Chao & Tung, 2001; Cao et al., 2005; Chen & Zhao, 2011; Chen et al., 2011). The  
19 lower number of studies focusing on indoor in comparison to outdoor air is generally  
20 attributed to the complexity in the determination of PM levels indoors, given the broad  
21 variability of direct indoor sources (printer emissions, dust re-suspension, smoking, cleaning,  
22 etc.) as well as the multiplicity of microenvironments (office spaces, industrial facilities,  
23 households, etc.). Evidently, particle levels in indoor environments are greatly influenced by  
24 indoor activities, which are characterised by typically generating high-concentration but short-  
25 term pollution events (acute events; Long et al., 2000). The examination of short-term  
26 variations in particulate concentrations is, therefore, essential in indoor air quality studies. In  
27 addition, indoor levels of UFPs may also be greatly influenced by outdoor sources as a  
28 function of building and ventilation characteristics and habits (Morawska et al., 2001; Lunden  
29 et al., 2008; Martuzevicius et al., 2008; McNabola et al., 2011), by three major mechanisms:  
30 mechanical ventilation, natural ventilation, and infiltration. Three parameters are generally  
31 used to monitor the presence of outdoor particles in the indoor environment: the  
32 indoor/outdoor (I/O) ratio, the penetration factor and the infiltration factor (Chen & Zhao,  
33 2011). The major outdoor emission source of fine and ultrafine particles is vehicular traffic,  
34 which is considered a global public health problem (World Bank 2006) and one of the major  
35 sources of particulate matter in urban areas worldwide, and in particular in Barcelona (Amato  
36 et al., 2009).

1 In this respect, the influence of outdoor PM sources on indoor environments is a key issue to  
2 be taken into consideration. Over the past several decades, the influence of outdoor air  
3 pollutants in indoor environments is believed to have decreased due to a variety of factors  
4 (Heinrich, 2010), including the construction of more tightly sealed buildings and reduced  
5 ventilation rates to save energy. However, Morawaska et al. (2001) studied the  
6 indoor/outdoor (I/O) relationships in the residential environment in Australia, and found ratios  
7 varying from 0.2 to 2.5 and depending on particle size distribution. Other studies have been  
8 conducted (Quackenboss et al., 1989; Wallace, 1996), which indicate that the ratio of the  
9 indoor to outdoor particle mass concentrations varies in a wide range from 0.5 to 2 in the  
10 absence of indoor particle sources. Conversely, in the presence of indoor sources such as  
11 smoking or other indoor combustion sources (e.g., fireplaces), I/O ratios may be as high as  
12 31 (Chen & Zhao, 2011). More recently, I/O relationships have been studied in the US and  
13 Australia with a special focus on the penetration rates of traffic-related particulates (Geller et  
14 al., 2002; Baxter et al., 2008; Lunden et al., 2008; Martuzevicius et al., 2008; Morawska et  
15 al., 2009). In Asia, I/O ratios were found to vary considerably due to the difference in size-  
16 dependent indoor particle emission rates, the geometry of the cracks in building envelopes,  
17 and the air exchange rates, thus making it difficult to understand the indoor/outdoor  
18 relationship using the I/O ratio only (Chen & Zhao, 2011). As for Europe, fewer studies are  
19 available and they focus mainly on modelling indoor air quality in indoor environments  
20 (Diapouli et al., 2007; Bohlin et al., 2008; Orosa & Baaliña, 2008), and on indoor air quality  
21 assessments in schools (Poupard et al., 2005; Fromme et al., 2007; Almeida et al., 2010).  
22 Studies on indoor air quality in workplace environments are especially rare in Southern-  
23 European countries, which are of special interest due to the influence of the Mediterranean  
24 climate on building ventilation systems (frequent air conditioning and natural ventilation by  
25 open windows).

26  
27 While most of the available studies focus on particle mass, there is still very little information  
28 on the indoor/outdoor relationship for other parameters such as particle composition,  
29 emission sources, particle number or surface area. However, UFPs are enriched in organic  
30 carbon content as well as prooxidative polycyclic aromatic hydrocarbons (PAH) that promote  
31 oxidative stress and inflammation, suggesting that it is not only the particle mass but also  
32 particle composition (in particular, carbon content) which should be considered when  
33 assessing indoor air quality and potential health effects. Understanding the relationship  
34 between outdoor and indoor aerosol particles for parameters other than PM mass is of  
35 importance to develop efficient control strategies to reduce health risk.

Based on the above considerations, the main objective of the present work is to understand the role of outdoor and indoor emission sources as determinants of indoor air quality in an urban environment. Specifically, this work focuses on the identification and characterisation of outdoor and indoor sources of fine and ultrafine particles in an indoor environment (a workplace in a typical Mediterranean urban area), and on understanding the factors influencing particle infiltration from ambient air into indoor environments. Going beyond the state-of-the-art, indoor air quality was assessed in terms of nanoscaled particulate mass ( $<1$  micron,  $PM_{10}$ ) and black carbon (BC) concentrations.

## Materials and methods

Simultaneous monitoring of outdoor and indoor  $PM_{10}$  and BC concentrations was carried out at a chemical research facility (IDAEA-CSIC), in the urban area of Barcelona (NE Spain). The building dates back to 1967, and has undergone no renovations ever since. The indoor location was a vacant laboratory located on the 5<sup>th</sup> floor, adjacent to other laboratories and offices which were in use on a daily basis during the study period. The studied laboratory (vacant) had the same dimensions, geometry, windows and doors as the laboratories in use, and was connected to them by means of a corridor. The doors of the vacant laboratory were kept open at all times in order to ensure air mass circulation with the remaining offices and laboratories on the same floor (which were occupied at the time). Windows were kept closed at all times, and there was no air conditioning. Indoor BC and  $PM_{10}$  concentrations were compared with outdoor BC and  $PM_{10}$  levels from an urban background monitoring station located at  $<150$ m from the research facility. The background station was located in a park in the vicinity of one of the major traffic arteries in the city, with a mean flow of approximately 100.000 vehicles/day.

Two sets of identical instruments were deployed at the indoor and outdoor monitoring stations. Black carbon concentrations were monitored using two multi-angle absorption photometers (MAAP) with a 10-minute (indoor) and 1-hour (outdoor) time resolution. The noise of the MAAP has been determined to be  $0.22 \text{ Mm}^{-1}$  for 1 min averaging interval (Müller et al., 2010), and the uncertainty of the instrument is  $\pm 12\%$  (Schladitz et al., 2009).  $PM_{10}$  concentrations were monitored by means of two laser spectrometers (GRIMM 1107) with 15-minute (indoor) and 30-minute (outdoor) time resolutions. The laser spectrometers are based on single particle detection and counting system by laser light scattering at  $90^\circ$ , which classifies the particles by size distribution into 31 channels and generates as output  $PM_{10}$ ,  $PM_{2.5}$  and  $PM_{10}$  mass concentrations based on the corresponding specific density factors.

The measured particle size range is 0.25 to  $>32\ \mu\text{m}$  (according to the manufacturer), with a sampling efficiency  $>95\%$  (Bukowiecki et al., 2011). All instruments were serviced before the monitoring campaign. Both laser spectrometers are routinely calibrated by comparison with reference gravimetric high-volume samplers, and corrected by means of a factor. For the purpose of this study, one high-volume sampler was set up in the indoor environment in order to obtain the indoor-specific correction factor. The temporal resolution of the instruments located indoor was set higher than outdoor aiming to observe short-term emissions generated by indoor sources. Monitoring of BC and  $\text{PM}_{10}$  was carried out during a 4-month period between 22/03/2010 and 28/07/2010 (with breaks due to technical issues).

In addition to online monitoring of BC and  $\text{PM}_{10}$ ,  $\text{PM}_{2.5}$  aerosols were collected on filter substrates for chemical analysis. In the indoor environment, one low-volume  $\text{PM}_{2.5}$  sampler ( $1\ \text{m}^3/\text{h}$ ) collected samples on 47-mm quartz fibre filter substrates during the day (9:00-17:00h, local time) and night (18:00-8:00h) shifts. In total, 66 valid samples were collected. In the outdoor site,  $\text{PM}_{2.5}$  samples were collected over 24-hr periods by means of a high-volume sampler (MCV SA,  $30\ \text{m}^3/\text{h}$ ). A total of 30 valid samples was collected. The  $\text{PM}_{2.5}$  mass was determined by gravimetry after the filters (samples and blanks) had acclimatised at controlled temperature conditions for at least 24h. The chemical composition of the particulates collected on the filters (indoor and outdoor) was characterised by means of inductively-coupled plasma mass and atomic emission spectrometry (ICP-MS and ICP-AES, for major and trace elements), and ion chromatography (IC) and flow injection analysis (FIA) for water-soluble ions, according to the methodology described in Querol et al (2001). A detailed description of the uncertainties associated with the PM mass and chemical analyses is provided elsewhere (Viana et al., 2006; Minguillón et al., 2007). Finally, 8  $\text{PM}_{2.5}$  24-hr samples were collected in the indoor environment by means of a reference high-volume sampler. The aim was to compare the online  $\text{PM}_{2.5}$  data and the low-volume mass concentrations with those obtained with the reference high-volume sampler, and thus obtain correction factors for the non-reference instruments.

## Results and Discussion

### Indoor and outdoor BC and $\text{PM}_{10}$ concentrations

Mean, minimum and maximum levels of BC and  $\text{PM}_{10}$  at the indoor and outdoor locations are summarised in Table 1. Indoor and outdoor  $\text{PM}_{10}$  levels, determined by laser spectrometry, were corrected with respect to the reference gravimetric method by applying the same

correction factor obtained for  $PM_{2.5}$  (indoor  $r^2=0.86$ ;  $y=0.67x+2.24$ ; outdoor  $r^2=0.77$ ;  $y=0.88x-0.63$ , Figure 1).

On average, BC and  $PM_{10}$  levels were higher in the outdoor environment than indoors (19% and 43% higher, respectively), with mean levels for the entire monitoring period of 1572  $ngBC/m^3$  and 7.6  $\mu gPM_{10}/m^3$  indoors, and 1871  $ngBC/m^3$  and 12.5  $\mu gPM_{10}/m^3$  outdoors. Maximum BC concentrations (15- or 30-minute means) were, however, higher in the indoor environment (7% higher), as a result of the peak events described above. Mean indoor/outdoor (I/O) ratios for the entire monitoring period (calculated on a sample-by-sample basis) were 0.97 and 0.76 for BC and  $PM_{10}$ , respectively, which are higher than values found in the literature for European urban environments for BC (0.60-0.65, Lunden et al., 2008), and also higher than average for  $PM_{10}$  but within the ranges described in the literature (0.14-1.37 for particles with aerodynamic diameter between 0.5 and 1.5  $\mu m$ , Chen & Zhao, 2011). Out of the 17 studies reviewed in the work by Chen & Zhao (2011) on I/O ratios for particles between 0.5 $\mu m$  and 1.5 $\mu m$ , 60% (10 studies) reported I/O ratios <0.75. Thus, the higher I/O ratios obtained for  $PM_{10}$  and BC in the present study evidence a major influence from outdoor particles in the indoor environment (especially BC), probably as a result of inadequate insulation of the building due to its old age (>40 years). Structural characteristics of the buildings such as insulation materials or ventilation patterns have been shown to play a more important role in affecting the indoor concentrations of the traffic-related aerosol, than parameters such as the distance to major roads or traffic density (Lunden et al., 2008; Martuzevicius et al., 2008). The ratios were calculated on a sample-by-sample basis, and then averaged (not calculated as the ratio of the means).

The time series of simultaneous indoor and outdoor  $PM_{10}$  and BC concentrations were analysed in search for emission patterns. As shown in Figure 2, the correlation between indoor and outdoor measurements was significantly high, with good correspondence in absolute values ( $y=1.07x$ ) although with a relatively high dispersion ( $r^2=0.35$ ) in the case of BC. The results evidenced a clear daily pattern coinciding with vehicular traffic emissions, with maximum levels during traffic rush hours, especially visible for BC. The strong correlation between the indoor and outdoor levels of both parameters, as well as the similarity in the absolute concentrations, suggests vehicular traffic as the dominant and common emission source in both environments.

#### Infiltration of outdoor emissions

1 In order to quantify the influence indoors of outdoor  $PM_{10}$  and BC emissions, penetration  
2 factors were calculated (Kuhlbusch et al., 2009). The aim was to determine the infiltration  
3 rate of outdoor particles into the indoor environment, in order to be able to: a) discriminate  
4 between the indoor particle emissions and the background levels of outdoor origin, and b)  
5 quantify the contributions from each of these sources in terms of  $PM_{10}$  and BC. Outdoor-to-  
6 indoor ratios (O/I) were calculated for time periods during which no indoor PM emissions  
7 were generated, typically weekends or Sundays in industrial facilities and workplaces. In the  
8 facility under study, weekends were representative of periods with zero indoor-generated  
9 emissions. The O/I ratio for BC (Table 2, calculated for the 3 weekends for which  
10 simultaneous indoor and outdoor BC data were available) showed limited variation between  
11 May and July (spring vs. summer), with a mean value of  $1.29 \pm 0.08$  (O/I = 1.20 to 1.35).  
12 Thus, the infiltration of BC particles was relatively constant and showed no seasonal  
13 dependence. Conversely, the O/I ratios for  $PM_{10}$  (Table 2, 7 weekends available) showed a  
14 statistically significant decrease of 42% from April to July at a 0.01 confidence level  
15 according to the Mann Kendall test (Salmi et al., 2002). Whereas the mean O/I ratio for  $PM_{10}$   
16 was 2.13 in April, it was closer to 1 (1.14-1.23) in July, indicating a much higher degree of  
17 particle infiltration during summer than in spring. Given that windows remained closed at all  
18 times, this increase in particle infiltration was interpreted as a change in particle  
19 characteristics, either particle composition and/or size.

20  
21 One possible interpretation for the different trends detected for the O/I ratios for BC and  $PM_{10}$   
22 could be the faster summer oxidation rates of  $SO_2$  due to enhanced photochemistry, which  
23 results in larger contributions to  $PM_{10}$  from sulphate particles with a finer grain size distribution  
24 and thus an enhanced infiltration from outdoors to the indoor environment. This process  
25 would not influence the penetration ratio of BC particles, with a grain size distribution which is  
26 unaffected by seasonal trends. In order to test this hypothesis, the sulphate and nitrate  
27 content of the indoor  $PM_{2.5}$  aerosols was analysed (Figure 3), assuming a similar distribution  
28 in  $PM_{10}$  based on the fine grain size distribution of atmospheric sulphate (Seinfeld & Pandis,  
29 1998). Results showed a marked increase in the sulphate content of outdoor  $PM_{2.5}$  aerosols,  
30 which accounted for 8% of  $PM_{2.5}$  in March and increased progressively to 29% in July. At the  
31 same time, ambient levels of nitrate decreased from 14% (March) to 2% (July) in the  $PM_{2.5}$   
32 fraction as a result of the thermal instability of this species. In parallel to the trends observed  
33 in the outdoor environment, sulphate levels in indoor  $PM_{2.5}$  (and thus in  $PM_{10}$ ) increased  
34 between March (10% of  $PM_{2.5}$ ) and June (22%), while the contribution from coarser nitrate  
35 particles decreased (from 19% to 4%). Thus, results evidenced the seasonal variability of the  
36 chemical composition of outdoor particulates, which implied a shift to a finer grain size



distribution and thus resulted in higher infiltration rates into the indoor environment. Consequently, the chemical composition of indoor particles was also seasonally dependent.

In addition to this, it is worthy to note that the indoor-outdoor temperature differences cause two-way air flows and thus lead to a significant influence on particle and pollutant penetration or infiltration (Chen et al., 2011a,b). Consequently, the seasonality of the indoor-outdoor temperature differences should also affect the infiltration of outdoor particles in the indoor environment. In the present work air exchange ratios were not measured due to the fact that there was no direct mechanical or manual ventilation in the study laboratory, given that one of the main goals of the study was to assess the impact of the building envelope on indoor particle levels.

In conclusion, particle infiltration does not only depend on physical barriers (e.g., windows, insulation) determining the outdoor-indoor exchange of particulates, but also on the physico-chemical properties of those particles. Therefore, in order to accurately discriminate the contribution to indoor air quality of particulates originating from indoor and outdoor emissions, it is necessary to monitor the variability of the calculated penetration factors as a function of time. This is not the case for studies in industrial facilities, e.g., aiming to detect leaks in manufacturing processes, for which point measurements of penetration ratios (e.g., one weekend) are recommended (Kuhlbusch et al., 2009). Our results evidence that, when infiltration of outdoor particulates is a major source of indoor aerosols, the variability of penetration factors should be monitored over time.

Indoor BC and  $PM_{10}$  emissions were estimated based on the penetration factors described above and the outdoor and indoor BC and  $PM_{10}$  levels. Figure 4 shows an example of the indoor BC concentrations monitored (total), as well as the calculated indoor BC of outdoor origin and indoor BC of indoor origin, for the month of July (when simultaneous indoor and outdoor BC data were available). Results showed that, on average and for the entire monitoring period, 70% of the indoor BC was originated outdoors, and 30% was generated by indoor emissions. The influence of outdoor emissions was only slightly higher for  $PM_{10}$ , for which 73% of the indoor mass originated from outdoor sources. These values are much higher than those reported by other authors (34% of indoor  $PM_{2.5}$  originating from outdoor traffic-derived sources, Martuzevicius et al., 2008). Even after the subtraction of the contribution from outdoor sources, indoor BC still showed peaks coinciding with vehicular traffic rush hours (Figures 4 and 5). This is a consequence of the application of mean weekend O/I ratios to hourly weekday data. Despite this artefact, this analysis allowed for the identification of indoor sources, as will be discussed below.



### Indoor BC and PM<sub>1</sub> emission sources

The daily cycle of total indoor BC (Figure 5, daily mean=1572 ng/m<sup>3</sup>) showed a clear traffic-derived trend, which contrasted with indoor BC emissions (daily mean=449 ng/m<sup>3</sup>) which were characterised by one major increase between 6:00-12:00 UTC (8:00-14:00 local time) and lower levels in the afternoon and evening hours. The morning increase was interpreted as the reflection of workplace activities linked to BC sources (printing, photocopying, etc), which were not located directly in the study laboratory and therefore their contributions to BC levels were not observed as the sharp, short-term BC peaks characteristic of printer emissions (He et al., 2010). Most probably, printing and copying activities were carried out during the morning, generating the BC increase shown in Figure 5. These activities were not dominant after the lunch break (13:00-14:00 local time). Other potential BC sources may be present but were not identified by this analysis. The potential contamination due to the monitoring instruments (pumps) was estimated in a maximum constant contribution of 250 ng/m<sup>3</sup> (Figure 4). As described above, the influence of outdoor traffic-derived aerosols was still detected as a secondary BC peak at 6:00-7:00 UTC (8:00-9:00 local time). This indicates that particles infiltrated from the outdoor environment cannot be fully segregated by applying O/I ratios in this type of work environment, even though this methodology is successfully applied in industrial workplaces. The reason for this is that particles are emitted at much higher concentrations in industrial environments, and they dominate over particulates infiltrated from ambient air.

Contrarily to the BC daily cycle, total indoor PM<sub>1</sub> levels showed relatively small hourly variations (mean daily PM<sub>1</sub> = 7.3 µg/m<sup>3</sup>, standard deviation = 0.32 µg/m<sup>3</sup>), which result from the sum of the indoor and outdoor daily emission trends. In ambient air, PM<sub>1</sub> levels decreased between 10:00 and 19:00 UTC (12:00-21:00 local time) mainly as a result of dilution due to the increased boundary layer height at midday and in the afternoon hours. As for the indoor PM<sub>1</sub> emissions (daily mean=1.8 µg/m<sup>3</sup>), their daily cycle was affected by the same artefact described for BC, and showed a slight impact of outdoor emissions at 7:00 UTC. Aside from this, PM<sub>1</sub> emissions peaked in parallel with those of BC between 8:00 and 12:00 UTC, probably as a result of worker passage along the corridor and between laboratories. Contrarily to BC, indoor PM<sub>1</sub> emissions remained relatively high (2-2.5 µg/m<sup>3</sup>) in the afternoon hours, only to decrease at the end of the shift (17:00 UTC). Consequently, indoor air quality was affected by BC and PM<sub>1</sub> levels during the morning hours (until the

lunch break), whereas in the afternoon hours and until the end of the shift indoor emissions were dominated by PM<sub>1</sub> sources.

### Work day vs. weekend analysis

Due to the artefact described above, when the daily cycle of indoor BC was analysed separately for work days and weekends (Figure 6), the morning traffic peak was detected throughout the entire week (even though with different intensities) given that traffic emissions decrease on weekends, but do not cease. As expected, the traffic peak was slightly delayed (1 hour) on weekends, with respect to work days. Conversely, the 8:00-12:00 UTC peak (due to workplace activities) was only present during weekdays, being absent on weekends when the building was empty. In terms of mass, during the 8:00-12:00 period, BC levels were on average 546 ng/m<sup>3</sup> higher on work days with respect to weekends (considered the background levels) due to workplace activities (61% increase, on average 346 ng/m<sup>3</sup> on weekends and 892 ng/m<sup>3</sup> on work days). This increase accounted for 25-30% of the total indoor BC concentrations during the maximum indoor emission period (8:00-12:00 UTC). Similar results were obtained for PM<sub>1</sub>, even though two unexplained indoor emission peaks were detected at 10:00-11:00 UTC and 15:00-16:00 UTC. Because of the extremely low variability of the levels of indoor PM<sub>1</sub> described in the previous section and Figure 5, these peaks could result from the uncertainty of the measurements. A rough estimation suggested that the increase of the indoor PM<sub>1</sub> levels as a result of indoor emissions could be 1.1 µg/m<sup>3</sup> (hourly mean, 15-20% of total indoor PM<sub>1</sub> during the maximum emission period). For BC, the results from this analysis confirm the interpretation of the indoor emission sources described in the previous sections, pointing towards printing and copying tasks as the major sources of indoor BC emissions.

### Shift analysis

Epidemiological studies (Tovalin-Ahumada & Whitehead, 2007; Zhang et al., 2009) evidence the relevance taking into account the duration and time of work-shifts when assessing indoor air quality levels and their potential impact on human health. This type of analysis is especially relevant in industrial facilities, in which production is independent of the time of day. In the present study, a separate analysis of indoor air pollutants was carried out for the day (6:00-17:00 UTC) and night (18:00-05:00 UTC) shifts. During the night shift only the cleaning crew was present inside the building. Given that outdoor vehicular traffic was the

main source of indoor BC (accounting on average for 70% of indoor BC), results showed that the levels of this pollutant in the indoor environment were the lowest during the night shift (on average 31% lower, 2008 ng/m<sup>3</sup> during the day shift and 1385 ng/m<sup>3</sup> during the night, Table 3). Because of the low hourly variability of indoor PM<sub>1</sub> levels shown in Figure 4, only minimal differences were observed between the day and night shifts (5%, 7.4 µg/m<sup>3</sup> vs. 7.0 µg/m<sup>3</sup>, respectively). In conclusion, BC levels were lower during the night than during the day shift, resulting from the combination of minimal indoor emissions and reduced ambient levels (and thus, low infiltration of BC particulates). PM<sub>1</sub> levels were rather constant mainly due to the relative stability of indoor PM<sub>1</sub> concentrations. As for PM<sub>1</sub> constituents, the mean indoor PM<sub>1</sub> chemical composition was dominated by mineral matter (7.2 µg/m<sup>3</sup>), probably as a result of dust re-suspension by worker passage. It is important to note that filter samples were only collected on work days (no weekend sampling), and therefore the PM<sub>1</sub> concentrations during the days with filter samples were higher than the average for the entire monitoring period (14.9 µg/m<sup>3</sup> for days with filter samples vs. 7.6 for the entire monitoring period, Table 1). On average, the contribution from secondary inorganic (sum of sulphate, nitrate and ammonium) and sea-salt aerosols to indoor air was similar in terms of mass (4.0 and 4.2 µg/m<sup>3</sup>, respectively). When comparing day and night shifts, a marked decrease was observed for the mineral matter content (9.4 µg/m<sup>3</sup> during the day to 5.8 µg/m<sup>3</sup> during the night), thus confirming that dust re-suspension by the passage of workers along the corridor and within the laboratories is the main source of indoor PM<sub>1</sub>. The sum of Na and Cl<sup>-</sup> (initially grouped as marine aerosol) also showed a clear decrease (6.1 to 3.7 µg/m<sup>3</sup>), indicating that the source of these components is not natural sea-salt only, and that significant contributions to the levels of Cl<sup>-</sup> are derived from indoor emissions (given that the study location is a chemical research facility). Finally, the levels of secondary inorganic aerosols remained relatively constant between day and night (4.2-4.9 µg/m<sup>3</sup>), given that the main source of these components in the indoor environment is infiltration from outdoor air.

## Conclusions

The sources and infiltration processes of nanoscaled particulates (BC and PM<sub>1</sub>) were determined in an indoor environment. A major impact of outdoor vehicular traffic emissions on indoor particulates was detected, which resulted from the inadequate insulation of the building. Even ensuring that windows were closed at all times, at least 70% of indoor BC and 73% of PM<sub>1</sub> concentrations originated from outdoor emissions. Outdoor to indoor infiltration of fine and ultrafine particles was higher for BC than for PM<sub>1</sub> (O/I = 1.29±0.08 vs. 1.95±0.38, calculated for “zero indoor emission” periods), probably due to its finer grain size distribution.

Particle infiltration showed no variation with time in the case of BC, but it showed a statistically significant decrease for PM<sub>1</sub> (O/I=2.13 to 1.23) from spring to summer. This was due to an increase in the contribution from fine grain-sized sulphate particles. We conclude that particle infiltration does not only depend on physical constraints but also on the physico-chemical properties of the particles.

The indoor emission sources of BC and PM<sub>1</sub> aerosols were also investigated. Printing and copying activities were identified as the main BC emission sources, and during their maximum emission period (10:00-14:00h) mean hourly BC levels increased by 71% with respect to the same period on weekends. Sources of PM<sub>1</sub> were mainly related to dust re-suspension by the passage of workers, occurring in the morning and afternoon hours (except the lunch break). Mean hourly indoor PM<sub>1</sub> levels increased by 15-20% as a result of indoor emissions. On average, the chemical composition of indoor PM<sub>1</sub> was dominated by mineral matter due to re-suspension processes.

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## Figure captions

Figure 1. Correlation between PM<sub>2.5</sub> concentrations obtained by means of high-volume filter samples and laser spectrometers (GRIMM) in the indoor and outdoor environments.

Figure 2. Mean hourly indoor and outdoor BC and PM<sub>1</sub> levels monitored between 17/07/2010 and 26/07/2010.

Figure 3. Seasonal evolution of the sulphate and nitrate contents of indoor and outdoor PM<sub>2.5</sub> aerosols.

Figure 4. Time series of total indoor BC concentrations (measured), outdoor BC infiltration (calculated) and indoor BC emissions (calculated).

Figure 5. Daily cycle of outdoor, total indoor and indoor emissions for BC (ng/m<sup>3</sup>) and PM<sub>1</sub> (µg/m<sup>3</sup>).

Figure 6. Indoor BC and PM<sub>1</sub> emissions during work days (Monday to Friday) and on weekends (Saturday-Sunday).

## Tables

Table 1. Mean, minimum and maximum levels of black carbon (BC) and PM<sub>1</sub> at the indoor and outdoor locations, for the entire monitoring period. I/O: indoor/outdoor ratio. Ratios calculated on a sample-by-sample basis, not as the ratio of the mean values.

<b>BC</b>	Mean	Min.	Max.
Indoor (ng/m <sup>3</sup> )	1572	178	10740
Outdoor (ng/m <sup>3</sup> )	1871	115	10060
I/O	0.97	0.14	3.42
<b>PM<sub>1</sub></b>			
Indoor (µg/m <sup>3</sup> )	7.6	1.9	55.1
Outdoor (µg/m <sup>3</sup> )	12.5	1.3	61.3
I/O	0.76	0.21	6.0

1 Table 2. BC and PM<sub>1</sub> penetration ratios. S.d.: standard deviation.

2

Dates (weekends)	Outdoor/Indoor ratios		
	Mean	Min.	Max.
BC			
1-2/05/2010	1.30	0.45	2.61
10-11/07/2010	1.20	0.38	2.30
24-25/07/2010	1.35	0.58	3.23
Mean BC O/I	1.29±0.08 (s.d.)		
PM <sub>1</sub>			
3-4/04/2010	2.13	0.98	3.57
15-16/05/2010	1.98	0.76	3.04
29-30/05/2010	1.75	1.00	3.23
12-13/06/2010	1.46	0.75	2.87
19-20/06/2010	1.34	0.44	2.85
10-11/07/2010	1.14	0.74	1.45
24-25/07/2010	1.23	0.72	1.65
Mean PM <sub>1</sub> O/I	1.95±0.38 (s.d.)		

3

4

1 Table 3. Mean chemical composition ( $\mu\text{m}^3$ ) of indoor PM<sub>2.5</sub> over 24-hr periods and during  
 2 the day (6:00-17:00 UTC) and night (18:00-5:00 UTC) shifts. SIA: secondary inorganic  
 3 aerosols (sum of sulphate, nitrate and ammonium).

4

	Daily mean	Day shift	Night shift
CO <sub>3</sub> <sup>2-</sup>	1.4	1.9	0.9
SiO <sub>2</sub>	2.9	3.8	2.5
Al <sub>2</sub> O <sub>3</sub>	1.0	1.3	0.8
Ca	0.9	1.2	0.6
K	0.5	0.6	0.5
Na	1.0	1.1	0.9
Mg	0.1	0.2	0.1
Fe	0.3	0.3	0.3
P <sub>2</sub> O <sub>5</sub>	0.1	0.2	0.1
SO <sub>4</sub> <sup>2-</sup>	2.2	2.1	2.7
NO <sub>3</sub> <sup>-</sup>	0.9	1.1	1.2
Cl	3.2	5.0	2.7
NH <sub>4</sub> <sup>+</sup>	0.9	1.0	1.0
BC	1.6	2.0	1.4
Mineral	7.2	9.4	5.8
NA+Cl <sup>-</sup> (marine)	4.2	6.1	3.7
SIA	4.0	4.2	4.9

5

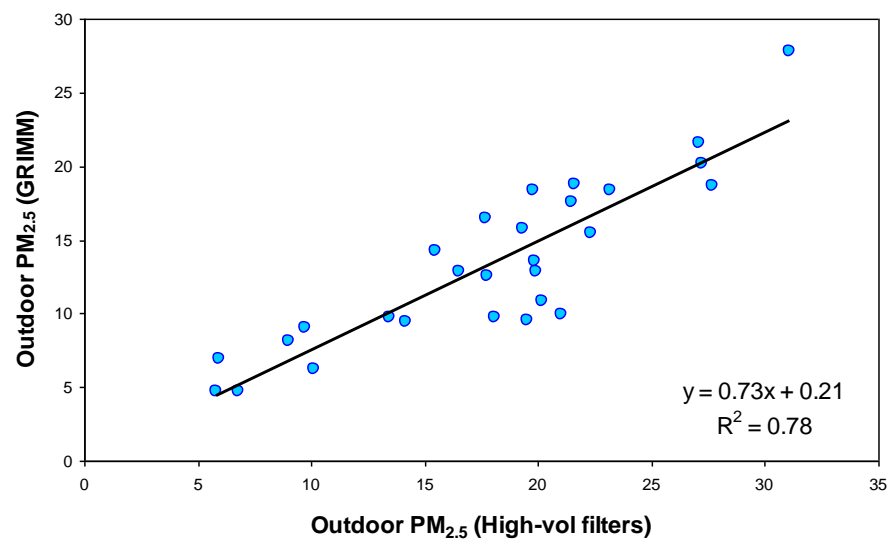
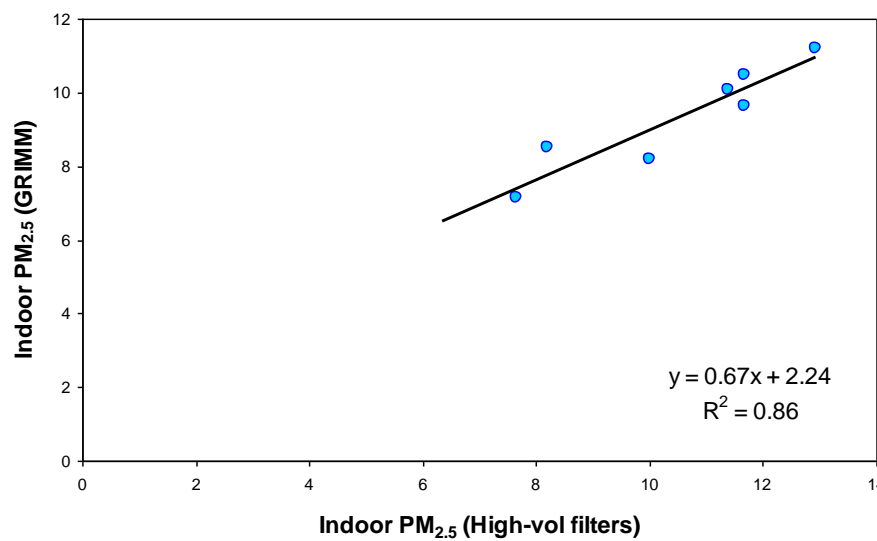


Figure 1

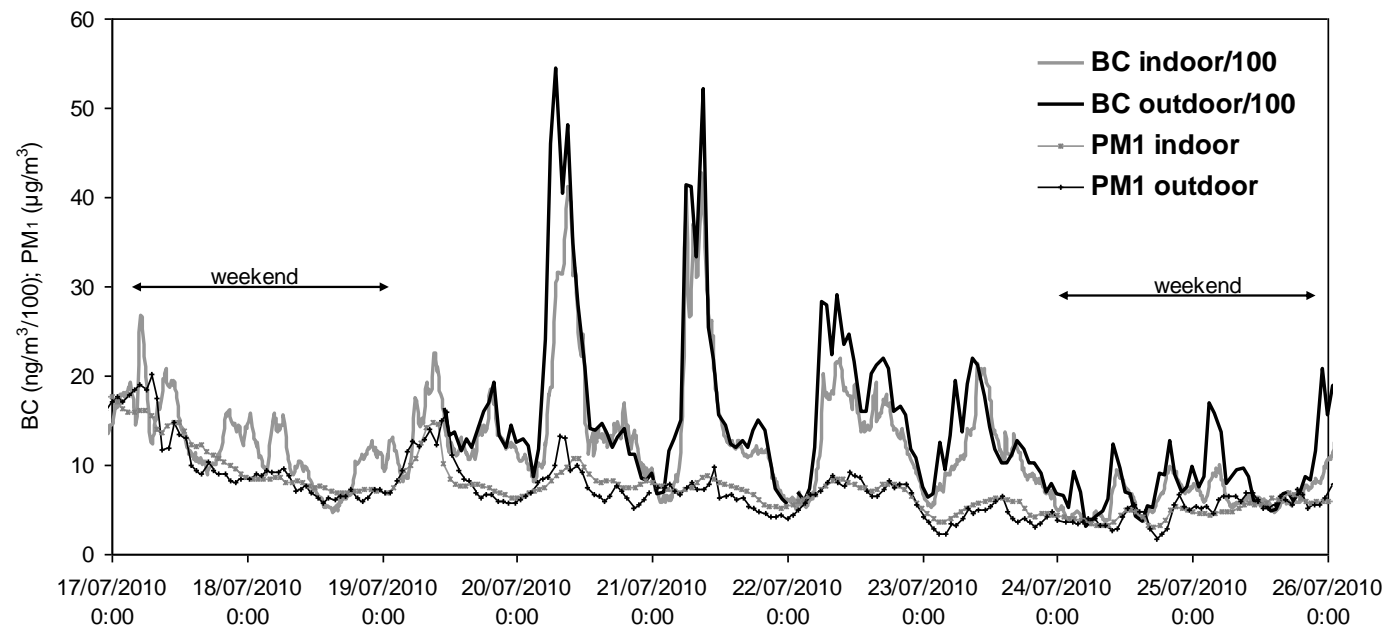


Figure 2

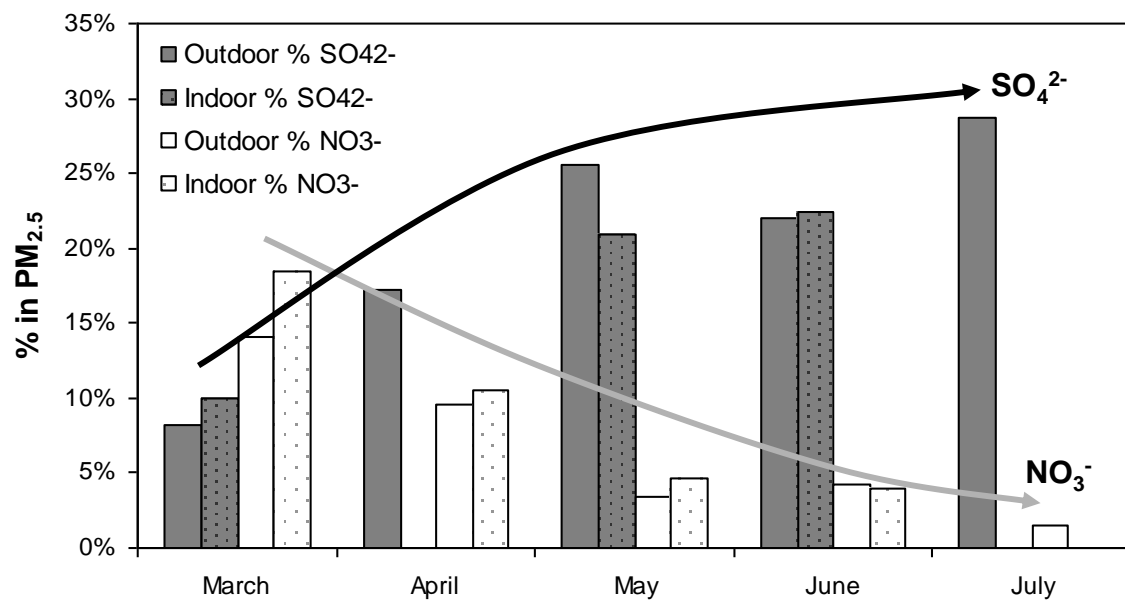


Figure 3



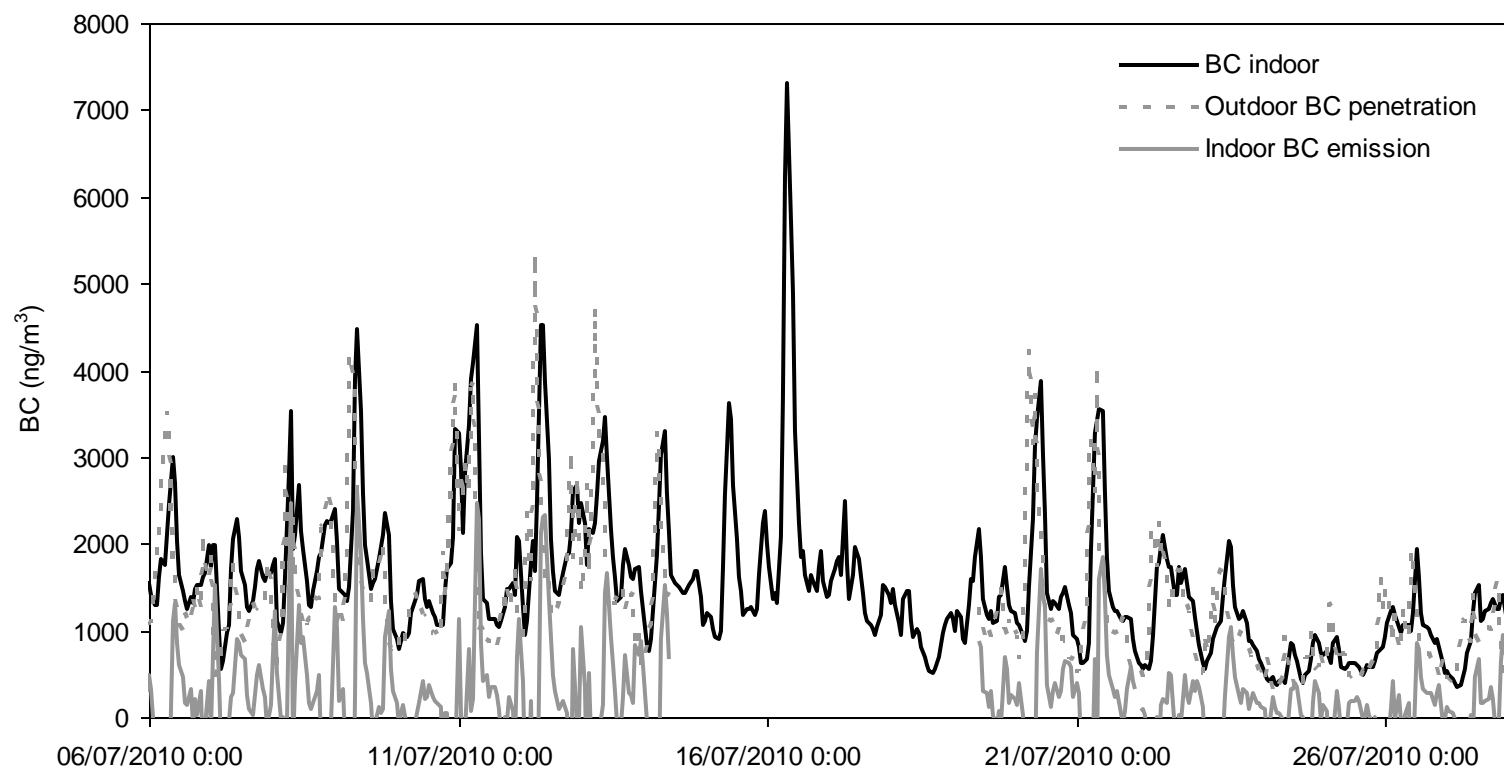


Figure 4

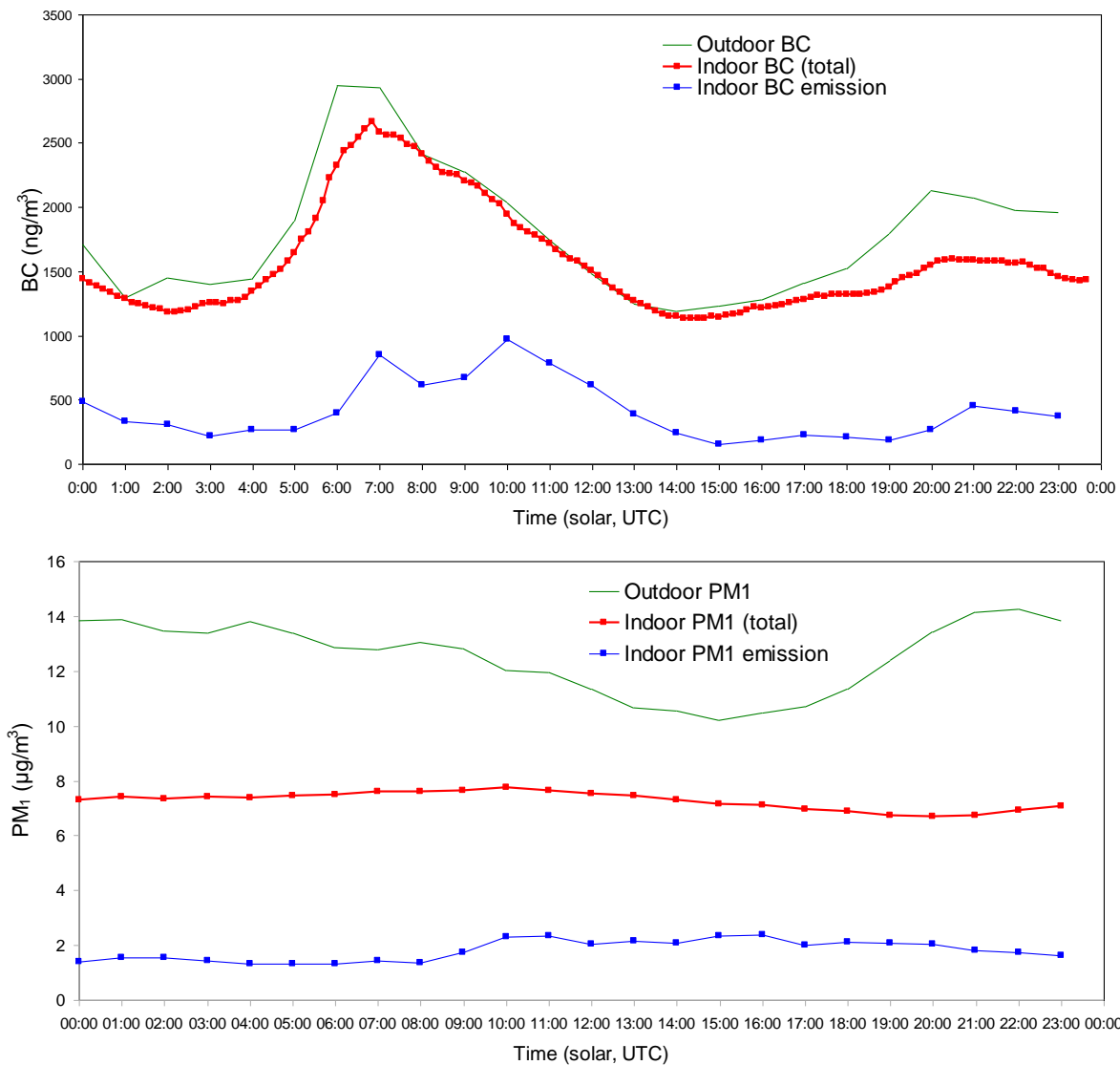


Figure 5

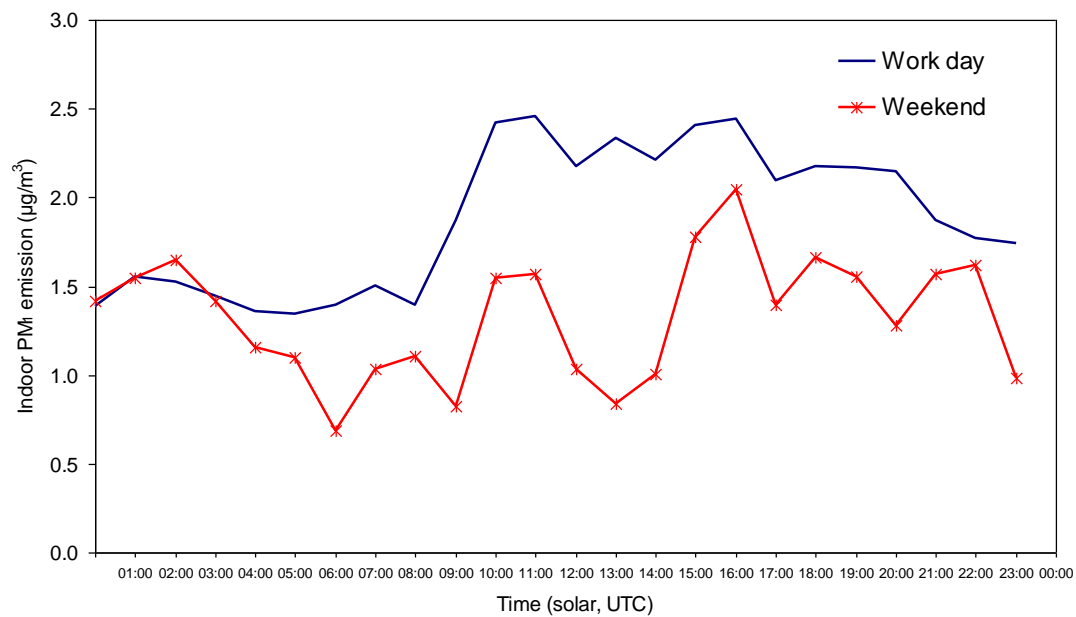
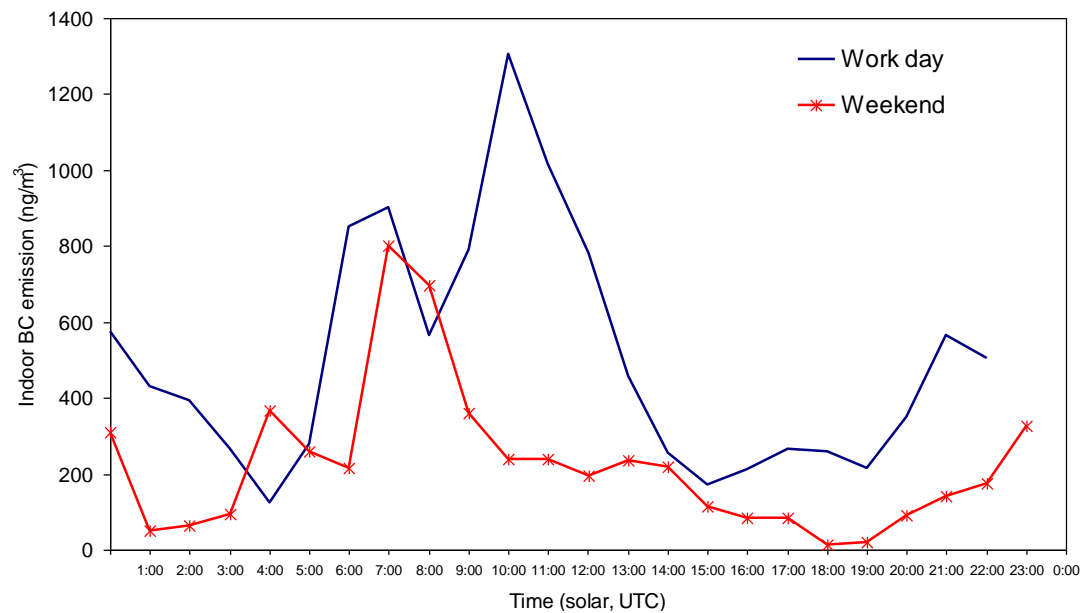


Figure 6